

# Nakajima-Zwanzig versus time-convolutionless master equation for the non-Markovian dynamics of a two-level system

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We consider the exact reduced dynamics of a two-level system coupled to a bosonic reservoir, further obtaining the exact time-convolutionless and Nakajima-Zwanzig non-Markovian equations of motion. The considered system includes the damped and undamped Jaynes-Cummings model. The result is obtained by exploiting an expression of quantum maps in terms of matrices and a simple relation between the time evolution map and time-convolutionless generator as well as Nakajima-Zwanzig memory kernel. This non-perturbative treatment shows that each operator contribution in Lindblad form appearing in the exact time-convolutionless master equation is multiplied by a different time dependent function. Similarly, in the Nakajima-Zwanzig master equation each such contribution is convoluted with a different memory kernel. It appears that depending on the state of the environment the operator structures of the two set of equations of motion can exhibit important differences.

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## I. INTRODUCTION

The study of open quantum systems is a wide research area of interest to various scientific communities, ranging from physicists to chemists and mathematicians. Its basic theoretical framework is well-understood when a Markovian description can be applied [1, 2], but despite important work [2, 3] a lot remains to be clarified and understood when considering non-Markovian dynamics, which is often mandatory for a realistic approach, also coping with strong coupling between system and environment. Recently a lot of research work has been devoted to the subject [4–24]. On the one hand it is not fully clear which is the most general operator structure of non-Markovian equations of motion, which do provide a well-defined time evolution and in particular preserves complete positivity. On the other hand one would like to link, in a possibly intuitive way, operator structures giving a sensible dynamical evolution with microscopic information on the physics of the system of interest. For the Markovian case both these approaches, phenomenological and microscopic, are well understood and successfully used. The result by Gorini, Kossakowski, Sudarshan and Lindblad [25, 26] provides a robust framework in order to envisage a possible ansatz for the evolution equations, while the Markov approximation is often connected with weak coupling, thus making microscopic approaches more manageable. In considering non-Markovian dynamics, there is a natural tendency to bring over the intuition gained for the Markovian case. As it appears, however, the non-Markovian case is much more subtle and involved, so that in such a way one often runs into incon-

sistencies and pitfalls.

In this article we will discuss a realistic physical model, simple enough to be exactly treated in detail, but already showing typical non-Markovian features. In particular we not only derive the exact equations of motion, which provide the counterpart to the Lindblad structure of a Markovian model, but also obtain a clear-cut connection to the microscopic approach via the two standard Nakajima-Zwanzig and time-convolutionless techniques. This work should help in better understanding typical non-Markovian features in simple but realistic models. We will consider a two-level system coupled first to a single mode of the radiation field, and later to a bath of harmonic oscillators, via a Jaynes-Cummings type of interaction. By exploiting the knowledge of the exact unitary evolution, and therefore of the reduced dynamics, as well as a suitable matrix representation of the dynamical maps, we can exhibit the exact time-convolutionless and Nakajima-Zwanzig equations of motion. It will appear that such equations are given by a sum of Lindblad terms, each multiplied or convoluted with a different time dependent function. Moreover the operator structure of these two sets of equations will be shown to be generally different, also depending on the environmental state. These exact results, with a clear-cut connection to a microscopic perturbative derivation, should guide in developing a correct intuition for the approximate treatment of more involved non-Markovian systems.

The article is organized as follows. In Sect. II we derive the map giving the exact reduced dynamics for the Jaynes-Cummings model, further introducing a simple representation of the map as a matrix with respect to a convenient basis of operators. In Sect. III we point out the simple relations connecting the time evolution map to the time-convolutionless, time-dependent generator and the Nakajima-Zwanzig memory kernel, respectively. These relationships are most easily expressed in

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matrix form. The corresponding matrices are evaluated for the reduced dynamics of our two-level system, further coming back to the operator expression of the master equations and giving the explicit result for a bath in the vacuum state. In Sect. IV we put forward a similar analysis for the damped two-level system, making contact with previous results and comparing with the undamped case.

## II. JAYNES-CUMMINGS MODEL AND EXACT REDUCED DYNAMICS

### A. Time evolution mapping

We consider a two-level system coupled to a single mode of the radiation field according to the total Hamiltonian

$$H = H_S + H_E + H_I, \quad (1)$$

where the system Hamiltonian is given by

$$H_S = \omega_0 \sigma_+ \sigma_-, \quad (2)$$

with  $\omega_0$  the transition frequency,  $\sigma_+ = |1\rangle\langle 0|$  and  $\sigma_- = |0\rangle\langle 1|$  the raising and lowering operators of the two-level system. The Hamiltonian for the single mode of the radiation field is given by

$$H_E = \omega b^\dagger b, \quad (3)$$

where the creation and annihilation operators  $b^\dagger$  and  $b$  obey the standard bosonic commutation relation. The coupling is in the Jaynes-Cummings form

$$H_I = g (\sigma_+ \otimes b + \sigma_- \otimes b^\dagger), \quad (4)$$

so that the considered model can describe e.g. the interaction between a two-level atom and a mode of the radiation field in electric dipole and rotating wave approximation. Working in the interaction picture with respect to the free Hamiltonian  $H_S + H_E$ ,

$$H_I(t) = g (\sigma_+ \otimes b e^{i\Delta t} + \sigma_- \otimes b^\dagger e^{-i\Delta t}), \quad (5)$$

with

$$\Delta = \omega_0 - \omega \quad (6)$$

the detuning between the system and the field mode, it is possible to obtain the exact dynamics generated by the total Hamiltonian (see e.g. Puri [27]), and therefore the reduced dynamics of the two-level system. We express the result exhibiting the unitary evolution operator which in the basis  $\{|1\rangle, |0\rangle\}$  is given by the following matrix, whose entries are operators in the Fock space of the radiation field

$$U(t) = \begin{pmatrix} c(\hat{n}+1, t) & d(\hat{n}+1, t) b \\ -b^\dagger d^\dagger(\hat{n}+1, t) & c^\dagger(\hat{n}, t) \end{pmatrix}, \quad (7)$$

where the following operators have been introduced

$$c(\hat{n}, t) = e^{i\Delta t/2} \left[ \cos\left(\sqrt{\Delta^2 + 4g^2\hat{n}} \frac{t}{2}\right) - i\Delta \frac{\sin\left(\sqrt{\Delta^2 + 4g^2\hat{n}} \frac{t}{2}\right)}{\sqrt{\Delta^2 + 4g^2\hat{n}}} \right], \quad (8)$$

$$d(\hat{n}, t) = -ie^{i\Delta t/2} 2g \frac{\sin\left(\sqrt{\Delta^2 + 4g^2\hat{n}} \frac{t}{2}\right)}{\sqrt{\Delta^2 + 4g^2\hat{n}}}, \quad (9)$$

with  $\hat{n} = b^\dagger b$  the number operator. The unitarity of  $U(t)$  is granted because of the easily verified relation

$$c^\dagger(\hat{n}, t) c(\hat{n}, t) + \hat{n} d^\dagger(\hat{n}, t) d(\hat{n}, t) = 1. \quad (10)$$

Given the unitary evolution of the whole bipartite system one can obtain the reduced dynamics of the two-level atom simply by taking the partial trace with respect to the environmental degrees of freedom. If the initial state of system and environment is factorized, the map giving the reduced dynamics is completely positive and takes the form

$$\begin{aligned} \rho(t) &= \text{Tr}_E [U(t)\rho(0) \otimes \rho_E U^\dagger(t)] \\ &\equiv \Phi(t)\rho(0). \end{aligned} \quad (11)$$

Taking  $U(t)$  as in Eq. (7) and considering an environmental state commuting with the number operator,  $[\rho_E, n] = 0$ , so that in particular both the vacuum and a thermal state can be dealt with, one comes to the following explicit expression for the action of the map  $\Phi(t)$ :

$$\begin{aligned} \rho_{11}(t) &= \rho_{00}(0) (1 - \alpha(t)) + \rho_{11}(0) \beta(t), \\ \rho_{10}(t) &= \rho_{10}(0) \gamma(t), \\ \rho_{01}(t) &= \rho_{01}(0) \gamma^*(t), \\ \rho_{00}(t) &= \rho_{00}(0) \alpha(t) + \rho_{11}(0) (1 - \beta(t)). \end{aligned} \quad (12)$$

The effect of the interaction with the bath is contained in the time dependent coefficients  $\alpha(t)$ ,  $\beta(t)$  and  $\gamma(t)$ , which are given by the following expectation values over the state of the environment  $\rho_E$ :

$$\begin{aligned} \alpha(t) &= \langle c^\dagger(\hat{n}, t) c(\hat{n}, t) \rangle_E, \\ \beta(t) &= \langle c^\dagger(\hat{n}+1, t) c(\hat{n}+1, t) \rangle_E, \\ \gamma(t) &= \langle c(\hat{n}, t) c(\hat{n}+1, t) \rangle_E. \end{aligned} \quad (13)$$

### B. Matrix representation

Now that we have obtained the completely positive map  $\Phi(t)$  giving the exact reduced time evolution of the considered two-level system, we shall express it for later convenience in matrix form with respect to a suitable basis of operators in the Hilbert space  $\mathbb{C}^2$ , following [28]. Indeed, having fixed a basis  $\{X_l\}_{l=0,1,2,3}$  of operators in

$\mathbb{C}^2$ , orthonormal according to  $\text{Tr}_S [X_k^\dagger X_l] = \delta_{kl}$ , a linear map  $\Lambda$  can be expressed in this basis according to

$$\Lambda \rho = \sum_{kl} L_{kl} \text{Tr}_S [X_l^\dagger \rho] X_k, \quad (14)$$

where the matrix of coefficients  $L_{kl}$  uniquely associated to the map is given by

$$L_{kl} = \text{Tr}_S [X_k^\dagger \Lambda (X_l)]. \quad (15)$$

Here and in the following we will use Greek or calligraphic letters to denote maps, and Roman letters to indicate the corresponding matrix. The convenient choice of basis for our calculations, in order to later recast the relevant maps in operator form, is given by  $\{X_l\}_{l=0,i} \equiv \left\{ \frac{1}{\sqrt{2}} \mathbb{1}, \frac{1}{\sqrt{2}} \sigma_i \right\}$ , where  $\sigma_i$  denote the usual Pauli operators. This choice leads to the following expression for the matrix  $F_{kl}(t)$  associated to the time evolution map  $\Phi(t)$ :

$$F(t) = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & \gamma_R(t) & \gamma_I(t) & 0 \\ 0 & -\gamma_I(t) & \gamma_R(t) & 0 \\ \beta(t) - \alpha(t) & 0 & 0 & \beta(t) + \alpha(t) - 1 \end{pmatrix}, \quad (16)$$

where the coefficients defined in Eq. (13) appear, and we denote with  $R$  and  $I$  real and imaginary part of a given function:

$$\gamma = \gamma_R + i\gamma_I.$$

This strategy of associating matrices to maps, already pursued in [28], and which transforms composition in matrix multiplication, will turn out to be very convenient to obtain the expressions of exact equations of motion leading to the dynamics described by Eq. (12).

### III. EXACT NAKAJIMA-ZWANZIG AND TIME-CONVOLUTIONLESS MASTER EQUATIONS

#### A. General expression in terms of time evolution map

With the aid of the knowledge of the exact time evolution, and using the representation of maps in terms of

matrices, we will now explicitly obtain two kinds of exact equations of motion for the reduced system's dynamics.

We first consider a master equation in differential form with a generator local in time, i.e. the time-convolutionless master equation. Assuming the existence of such a generator  $\mathcal{K}_{\text{TCL}}(t)$ , it should obey the equation

$$\dot{\rho}(t) = \mathcal{K}_{\text{TCL}}(t)\rho(t), \quad (17)$$

which due to  $\rho(t) = \Phi(t)\rho(0)$  is satisfied upon identifying

$$\mathcal{K}_{\text{TCL}}(t) = \dot{\Phi}(t)\Phi^{-1}(t), \quad (18)$$

or in terms of matrices

$$K_{\text{TCL}}(t) = \dot{F}(t)F^{-1}(t), \quad (19)$$

holding provided the inverse does exist.

On a similar footing one can consider a master equation in integrodifferential form, given by a suitable memory kernel, corresponding to the Nakajima-Zwanzig master equation. In this case the memory kernel  $\mathcal{K}_{\text{NZ}}(t)$  should obey the convolution equation

$$\dot{\rho}(t) = (\mathcal{K}_{\text{NZ}} \star \rho)(t), \quad (20)$$

so that in view of Eq. (11) one has the relation

$$\hat{\mathcal{K}}_{\text{NZ}}(u) = u\mathbb{1} - \hat{\Phi}^{-1}(u), \quad (21)$$

where the hat denotes the Laplace transform, and therefore in matrix representation

$$\hat{K}_{\text{NZ}}(u) = u\mathbb{1} - \hat{F}^{-1}(u). \quad (22)$$

It immediately appears that, given the time evolution map from the relationships Eq. (19) and Eq. (22), one can directly obtain the generator of the master equation in time-convolutionless form, or the memory kernel for the Nakajima-Zwanzig form respectively, without resorting to the evaluation of the whole perturbative series, which of course leads to the same result [20]. Obviously, given the exact time evolution one does not need the equations of motion. Nevertheless an exact comprehensive study as feasible in this case allows, as we shall see, to point out general features, serving as a guide for phenomenological or approximate treatments.

Starting from Eq. (16) and Eq. (19) one obtains for the model of interest

$$K_{\text{TCL}}(t) = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & \text{Re} \left[ \frac{\dot{\gamma}(t)}{\gamma(t)} \right] & \text{Im} \left[ \frac{\dot{\gamma}(t)}{\gamma(t)} \right] & 0 \\ 0 & -\text{Im} \left[ \frac{\dot{\gamma}(t)}{\gamma(t)} \right] & \text{Re} \left[ \frac{\dot{\gamma}(t)}{\gamma(t)} \right] & 0 \\ \frac{[1-2\beta(t)]\dot{\alpha}(t) - [1-2\alpha(t)]\dot{\beta}(t)}{\beta(t) + \alpha(t) - 1} & 0 & 0 & \frac{\dot{\beta}(t) + \dot{\alpha}(t)}{\beta(t) + \alpha(t) - 1} \end{pmatrix}, \quad (23)$$

and the expression is well defined provided the determinant

$$\det F(t) = |\gamma(t)|^2 [\alpha(t) + \beta(t) - 1] \quad (24)$$

is different from zero.

On a similar footing one can consider the Laplace transform of Eq. (16), given by the matrix  $\hat{F}(u)$  with determinant

$$\det \hat{F}(u) = \left[ \widehat{\gamma_R}^2(u) + \widehat{\gamma_I}^2(u) \right] \left[ \frac{\hat{\alpha}(u) + \hat{\beta}(u)}{u} - \frac{1}{u^2} \right], \quad (25)$$

and using Eq. (22) one further obtains

$$\hat{K}_{\text{NZ}}(u) = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & u - \frac{\widehat{\gamma_R}(u)}{\widehat{\gamma_R}^2(u) + \widehat{\gamma_I}^2(u)} & \frac{\widehat{\gamma_I}(u)}{\widehat{\gamma_R}^2(u) + \widehat{\gamma_I}^2(u)} & 0 \\ 0 & -\frac{\widehat{\gamma_I}(u)}{\widehat{\gamma_R}^2(u) + \widehat{\gamma_I}^2(u)} & u - \frac{\widehat{\gamma_R}(u)}{\widehat{\gamma_R}^2(u) + \widehat{\gamma_I}^2(u)} & 0 \\ \frac{u^2 [\hat{\alpha}(u) - \hat{\beta}(u)]}{1 - u [\hat{\alpha}(u) + \hat{\beta}(u)]} & 0 & 0 & \frac{2u - u^2 [\hat{\alpha}(u) + \hat{\beta}(u)]}{1 - u [\hat{\alpha}(u) + \hat{\beta}(u)]} \end{pmatrix}, \quad (26)$$

which upon inverse Laplace transform provides the exact Nakajima-Zwanzig integral kernel. As it appears, working with the matrix representation has proved very convenient to easily obtain the maps fixing the differential and integrodifferential equations of motion for the model, given by Eq. (17) and Eq. (20) respectively, in terms of the completely positive time evolution map Eq. (12).

## B. Operator expression

We now recast the obtained maps, providing time local generator and memory kernel, in operator form, to better compare with previous work and appreciate the difference in the obtained expressions. This is easily done observing that a matrix of the form

$$A = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & E_r & E_i & 0 \\ 0 & -E_i & E_r & 0 \\ X & 0 & 0 & Y \end{pmatrix} \quad (27)$$

in the basis  $\left\{ \frac{1}{\sqrt{2}} \mathbb{1}, \frac{1}{\sqrt{2}} \sigma_i \right\}$  corresponds in operator form to the map

$$\begin{aligned} \mathcal{A}\rho &= iE_i [\sigma_+ \sigma_-, \rho] \\ &+ \frac{1}{2} (X - Y) \left[ \sigma_+ \rho \sigma_- - \frac{1}{2} \{ \sigma_- \sigma_+, \rho \} \right] \\ &- \frac{1}{2} (X + Y) \left[ \sigma_- \rho \sigma_+ - \frac{1}{2} \{ \sigma_+ \sigma_-, \rho \} \right] \\ &+ \frac{1}{4} (Y - 2E_r) [\sigma_z \rho \sigma_z - \rho], \end{aligned} \quad (28)$$

whose last term can be written in alternative ways according to the identities

$$\begin{aligned} \sigma_z \rho \sigma_z - \rho &= 4 \left[ \sigma_+ \sigma_- \rho \sigma_+ \sigma_- - \frac{1}{2} \{ \sigma_+ \sigma_-, \rho \} \right] \\ &= 4 \left[ \sigma_- \sigma_+ \rho \sigma_- \sigma_+ - \frac{1}{2} \{ \sigma_- \sigma_+, \rho \} \right]. \end{aligned} \quad (29)$$

Exploiting this result one obtains the exact time-convolutionless master equation describing the reduced dynamics of a two-level atom coupled according to the Jaynes-Cummings model to a single mode of the radiation field, which is of the form Eq. (17) with  $\mathcal{K}_{\text{TCL}}(t)$  given by

$$\begin{aligned} \mathcal{K}_{\text{TCL}}(t)\rho &= i \text{Im} \left[ \frac{\dot{\gamma}(t)}{\gamma(t)} \right] [\sigma_+ \sigma_-, \rho] \\ &+ \frac{[\alpha(t) - 1] \dot{\beta}(t) - \beta(t) \dot{\alpha}(t)}{\beta(t) + \alpha(t) - 1} \left[ \sigma_+ \rho \sigma_- - \frac{1}{2} \{ \sigma_- \sigma_+, \rho \} \right] \\ &+ \frac{[\beta(t) - 1] \dot{\alpha}(t) - \alpha(t) \dot{\beta}(t)}{\beta(t) + \alpha(t) - 1} \left[ \sigma_- \rho \sigma_+ - \frac{1}{2} \{ \sigma_+ \sigma_-, \rho \} \right] \\ &+ \frac{1}{4} \left\{ \frac{\dot{\beta}(t) + \dot{\alpha}(t)}{\beta(t) + \alpha(t) - 1} - 2 \text{Re} \left[ \frac{\dot{\gamma}(t)}{\gamma(t)} \right] \right\} [\sigma_z \rho \sigma_z - \rho]. \end{aligned} \quad (30)$$

In a similar way one has for the Laplace transform of the memory kernel  $\mathcal{K}_{\text{NZ}}(t)$ , appearing in the exact Nakajima-

Zwanzig master equation Eq. (20), the expression

$$\begin{aligned} \hat{\mathcal{K}}_{\text{NZ}}(u)\rho = & i \frac{\hat{\gamma}_I(u)}{\hat{\gamma}_R^2(u) + \hat{\gamma}_I^2(u)} [\sigma_+\sigma_-, \rho] \\ & + \frac{u[u\hat{\alpha}(u) - 1]}{1 - u[\hat{\alpha}(u) + \hat{\beta}(u)]} \left[ \sigma_+\rho\sigma_- - \frac{1}{2} \{\sigma_-\sigma_+, \rho\} \right] \\ & + \frac{u[u\hat{\beta}(u) - 1]}{1 - u[\hat{\alpha}(u) + \hat{\beta}(u)]} \left[ \sigma_-\rho\sigma_+ - \frac{1}{2} \{\sigma_+\sigma_-, \rho\} \right] \\ & + \frac{1}{4} \left\{ \frac{u^2[\hat{\alpha}(u) + \hat{\beta}(u)]}{1 - u[\hat{\alpha}(u) + \hat{\beta}(u)]} + 2 \frac{\hat{\gamma}_R(u)}{\hat{\gamma}_R^2(u) + \hat{\gamma}_I^2(u)} \right\} \\ & \times [\sigma_z\rho\sigma_z - \rho]. \quad (31) \end{aligned}$$

Despite being exact these expressions are quite cumbersome, since the functions given in Eq. (13), which together with their Laplace transform determine the structure of these operators, depend on the specific expression of the environmental state. It is therefore convenient to consider a specific choice, allowing for a more detailed evaluation.

### C. The vacuum case

If the radiation field is in the vacuum state, the functions given in Eq. (13) simplify considerably, since  $\alpha(t) \rightarrow 1$ , while  $\beta(t)$  becomes a function of  $\gamma(t)$  according to  $\beta(t) \rightarrow |\gamma(t)|^2$ . The function  $\gamma(t)$  for the vacuum case is given by the expression

$$G^1(t) = e^{i\Delta t/2} \left[ \cos\left(\frac{\Omega_1 t}{2}\right) - i \frac{\Delta}{\Omega_1} \sin\left(\frac{\Omega_1 t}{2}\right) \right], \quad (32)$$

where the superscript recalls that we have a single mode of the radiation field, while  $\Omega_1 = \sqrt{\Delta^2 + 4g^2}$ . These results for the vacuum case greatly simplify the expression of the obtained master equations, and inserted in Eq. (24) show that the time-convolutionless master equation off-resonance is always well defined.

Indeed the time-convolutionless master equation for the vacuum case simply reads

$$\begin{aligned} \mathcal{K}_{\text{TCL}}^{\text{vac}}(t)\rho = & +i \text{Im} \left[ \frac{\dot{G}^1(t)}{G^1(t)} \right] [\sigma_+\sigma_-, \rho] \\ & - 2 \text{Re} \left[ \frac{\dot{G}^1(t)}{G^1(t)} \right] \left[ \sigma_-\rho\sigma_+ - \frac{1}{2} \{\sigma_+\sigma_-, \rho\} \right], \quad (33) \end{aligned}$$

corresponding as expected to a Lindblad structure with time dependent coefficients. Exploiting Eq. (32) one has

the explicit expression

$$\begin{aligned} \mathcal{K}_{\text{TCL}}^{\text{vac}}(t)\rho = & -ig^2\Delta \frac{1 - \cos(\Omega_1 t)}{\Omega_1} \left[ \cos^2\left(\frac{\Omega_1 t}{2}\right) + \frac{\Delta^2}{\Omega_1^2} \sin^2\left(\frac{\Omega_1 t}{2}\right) \right]^{-1} \\ & \times [\sigma_+\sigma_-, \rho] \\ & + 2g^2 \frac{\sin(\Omega_1 t)}{\Omega_1} \left[ \cos^2\left(\frac{\Omega_1 t}{2}\right) + \frac{\Delta^2}{\Omega_1^2} \sin^2\left(\frac{\Omega_1 t}{2}\right) \right]^{-1} \\ & \times \left[ \sigma_-\rho\sigma_+ - \frac{1}{2} \{\sigma_+\sigma_-, \rho\} \right], \quad (34) \end{aligned}$$

where in particular one directly sees that the coefficient in front of the dissipative term at the r.h.s. of Eq. (34) periodically takes on negative values, so that it describes a truly non-Markovian behavior and a Markovian approximation is not justified even for long times.

The choice of the vacuum as bath state brings in important simplifications also for the expression of the Nakajima-Zwanzig memory kernel, whose Laplace transform reads

$$\begin{aligned} \hat{\mathcal{K}}_{\text{NZ}}^{\text{vac}}(u)\rho = & +i \frac{\hat{G}_I^1(u)}{\hat{G}_R^1(u) + \hat{G}_I^1(u)} [\sigma_+\sigma_-, \rho] \\ & + \left[ \frac{1 - u\hat{z}^1(u)}{\hat{z}^1(u)} \right] \left[ \sigma_-\rho\sigma_+ - \frac{1}{2} \{\sigma_+\sigma_-, \rho\} \right] \\ & - \frac{1}{4} \left[ \frac{1 - u\hat{z}^1(u)}{\hat{z}^1(u)} + 2 \left( u - \frac{\hat{G}_R^1(u)}{\hat{G}_R^1(u) + \hat{G}_I^1(u)} \right) \right] \\ & \times [\sigma_z\rho\sigma_z - \rho], \quad (35) \end{aligned}$$

where

$$z^1(t) = |G^1(t)|^2. \quad (36)$$

Taking into account the explicit result Eq. (32) one obtains after some calculations the following compact expression for the memory kernel

$$\begin{aligned} \mathcal{K}_{\text{NZ}}^{\text{vac}}(\tau)\rho = & -ig^2 \sin(\Delta\tau) [\sigma_+\sigma_-, \rho] \\ & + 2g^2 \cos\left(\sqrt{\Delta^2 + 2g^2}\tau\right) \left[ \sigma_-\rho\sigma_+ - \frac{1}{2} \{\sigma_+\sigma_-, \rho\} \right] \\ & - \frac{1}{2}g^2 \left[ \cos\left(\sqrt{\Delta^2 + 2g^2}\tau\right) - \cos(\Delta\tau) \right] [\sigma_z\rho\sigma_z - \rho], \quad (37) \end{aligned}$$

which is always well-defined even on-resonance. These results already allow for a few important remarks. We first notice that the different operator contributions in Lindblad form appearing in the various time local and integral kernels are multiplied by different time dependent functions. This lesson, already learnt in other models [15, 20, 29, 30], tells us that as a general rule non-Markovian dynamics cannot be obtained from Markovian expressions by simply taking the convolution with

a single integral kernel, or multiplying by a single time dependent function. This fact has often been overlooked when seeking non-Markovian integrodifferential master equations [31–34], sometimes leading to unphysical behaviors. More than this, for the same model different sets of non-Markovian equations of motion can have different operator structures, as it appears comparing e.g. the time-convolutionless and Nakajima-Zwanzig results for the vacuum Eq. (33) and Eq. (37). This fact has already been noticed in [20], but the present analysis shows that this asymmetry depends on the choice of environmental state. For the present model it only appears in connection with the vacuum state. Indeed, while the disappearance of the term corresponding to excitation of the two-level system is obvious on physical grounds, when considering as bath state the vacuum, the vanishing of the coefficient in front of the dephasing term  $\sigma_z \rho \sigma_z - \rho$  is a peculiar feature of the time-convolutionless master equation.

#### IV. DAMPED TWO-LEVEL SYSTEM

##### A. Exact master equations for bath in the vacuum state

The technique used in Sect. III to obtain the time-convolutionless and Nakajima-Zwanzig equation of motions for a model whose evolution is known, by exploiting the representation of maps in terms of matrices, is applicable for a detailed study of simple models. We exploit it now for a variant of the model considered in Sect. II, in which the environmental Hamiltonian is a collection of harmonic oscillators

$$H_E = \sum_k \omega_k b_k^\dagger b_k, \quad (38)$$

and the interaction Hamiltonian is replaced by

$$H_I = \sum_k \left( g_k \sigma_+ \otimes b_k + g_k^* \sigma_- \otimes b_k^\dagger \right). \quad (39)$$

This model corresponds for a Lorentzian spectral density to the damped Jaynes-Cummings model. The time evolution map for this model, considering the special case of an environment in the vacuum state, has been obtained in [35] and can be expressed as:

$$\begin{aligned} \rho_{11}(t) &= \rho_{11}(0) |G(t)|^2, \\ \rho_{10}(t) &= \rho_{10}(0) G(t), \\ \rho_{01}(t) &= \rho_{01}(0) G^*(t), \\ \rho_{00}(t) &= \rho_{00}(0) + \rho_{11}(0) (1 - |G(t)|^2), \end{aligned} \quad (40)$$

where  $\rho(t) = \Phi^{\text{DVac}}(t)\rho(0)$ , since we are considering the damped model with the bath in the vacuum state. The function  $G(t)$  is the solution of the equation

$$\frac{d}{dt}G(t) = - \int_0^t dt_1 f(t-t_1) G(t_1) \quad G(0) = 1, \quad (41)$$

with  $f(t)$  the two-point correlation function given by

$$\begin{aligned} f(t-t_1) &= e^{i\omega_0(t-t_1)} \langle 0 | \sum_k g_k b_k e^{-i\omega_k t} \sum_j g_j^* b_j^\dagger e^{i\omega_j t_1} | 0 \rangle \\ &= \sum_k |g_k|^2 e^{i(\omega_0 - \omega_k)(t-t_1)}, \end{aligned} \quad (42)$$

corresponding to the Fourier transform of the spectral density.

Starting from Eq. (40) and exploiting the same strategy used in Sect. III one immediately obtains for the matrix representation of the time-convolutionless generator the expression

$$K_{\text{TCL}}^{\text{DVac}}(t) = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & \text{Re} \left[ \frac{\dot{G}(t)}{G(t)} \right] & \text{Im} \left[ \frac{\dot{G}(t)}{G(t)} \right] & 0 \\ 0 & -\text{Im} \left[ \frac{\dot{G}(t)}{G(t)} \right] & \text{Re} \left[ \frac{\dot{G}(t)}{G(t)} \right] & 0 \\ 2 \text{Re} \left[ \frac{\dot{G}(t)}{G(t)} \right] & 0 & 0 & 2 \text{Re} \left[ \frac{\dot{G}(t)}{G(t)} \right] \end{pmatrix}, \quad (43)$$

so that the master equation in operator form reads

$$\begin{aligned} \mathcal{K}_{\text{TCL}}^{\text{DVac}}(t)\rho &= +i \text{Im} \left[ \frac{\dot{G}(t)}{G(t)} \right] [\sigma_+ \sigma_-, \rho] \\ &\quad - 2 \text{Re} \left[ \frac{\dot{G}(t)}{G(t)} \right] \left[ \sigma_- \rho \sigma_+ - \frac{1}{2} \{ \sigma_+ \sigma_-, \rho \} \right], \end{aligned} \quad (44)$$

which confirms the result obtained in [2]. One can also determine the expression of the Nakajima-Zwanzig memory kernel, whose Laplace transform is given by

$$\hat{K}_{\text{NZ}}^{\text{DVac}}(u) = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & u - \frac{\widehat{G}_R(u)}{\widehat{G}_R^2(u) + \widehat{G}_I^2(u)} & \frac{\widehat{G}_I(u)}{\widehat{G}_R^2(u) + \widehat{G}_I^2(u)} & 0 \\ 0 & -\frac{\widehat{G}_I(u)}{\widehat{G}_R^2(u) + \widehat{G}_I^2(u)} & u - \frac{\widehat{G}_R(u)}{\widehat{G}_R^2(u) + \widehat{G}_I^2(u)} & 0 \\ \frac{u\hat{z}(u)-1}{\hat{z}(u)} & 0 & 0 & \frac{u\hat{z}(u)-1}{\hat{z}(u)} \end{pmatrix}, \quad (45)$$

where we have used the notation

$$z(t) = |G(t)|^2, \quad (46)$$

leading to the master equation

$$\begin{aligned} \hat{\mathcal{K}}_{\text{NZ}}^{\text{Vac}}(u)\rho = & +i \frac{\widehat{G}_I(u)}{\widehat{G}_R^2(u) + \widehat{G}_I^2(u)} [\sigma_+ \sigma_-, \rho] \\ & + \left[ \frac{1 - u\hat{z}(u)}{\hat{z}(u)} \right] \left[ \sigma_- \rho \sigma_+ - \frac{1}{2} \{ \sigma_+ \sigma_-, \rho \} \right] \\ & - \frac{1}{4} \left[ \frac{1 - u\hat{z}(u)}{\hat{z}(u)} + 2 \left( u - \frac{\widehat{G}_R(u)}{\widehat{G}_R^2(u) + \widehat{G}_I^2(u)} \right) \right] \\ & \times [\sigma_z \rho \sigma_z - \rho]. \end{aligned} \quad (47)$$

## B. Role of correlation function

We now show that this constructive approach leads to the same result as obtained by a suitable ansatz in [20], where the detailed connection with the perturbative expansion via projection operator techniques has also been considered. We first have to recall the relation in Laplace transform between the functions  $G(t)$  and  $f(t)$ . Indeed, for the case of a two-level system interacting with an environment of oscillators in the vacuum state, all functions appearing in the non-Markovian equations of motion can be related to a single correlation function of the model, given by Eq. (42).

To fully exploit this fact in order to express the memory kernel in the most compact and transparent way let us observe that according to Eq. (41) one has

$$\hat{f}(u) = \frac{\hat{G}^*(u)}{|\hat{G}(u)|^2} - u, \quad (48)$$

so that in view of the fact that for real  $u$

$$\text{Re} [\hat{h}(u)] = \widehat{\text{Re}} h(u) \quad (49)$$

together with

$$\text{Im} [\hat{h}(u)] = \widehat{\text{Im}} h(u), \quad (50)$$

the functions

$$\hat{f}_I(u) \text{ and } -\frac{\widehat{G}_I(u)}{\widehat{G}_R^2(u) + \widehat{G}_I^2(u)} \quad (51)$$

do coincide on the real axis, and therefore due to the identity principle on the common region of analyticity. A corresponding result holds for the functions

$$\widehat{f}_R(u) \text{ and } \frac{\widehat{G}_R(u)}{\widehat{G}_R^2(u) + \widehat{G}_I^2(u)} - u, \quad (52)$$

so that the matrix representing the Nakajima-Zwanzig kernel can be rewritten in a more compact way as

$$\hat{K}_{\text{NZ}}^{\text{DVac}}(u) = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & -\widehat{f}_R(u) & -\widehat{f}_I(u) & 0 \\ 0 & \widehat{f}_I(u) & -\widehat{f}_R(u) & 0 \\ \frac{u\hat{z}(u)-1}{\hat{z}(u)} & 0 & 0 & \frac{u\hat{z}(u)-1}{\hat{z}(u)} \end{pmatrix}, \quad (53)$$

leading indeed to the integral kernel first obtained in [20]

$$\begin{aligned} \mathcal{K}_{\text{NZ}}^{\text{DVac}}(\tau) \rho = & -i f_I(\tau) [\sigma_+ \sigma_-, \rho] \\ & + k_1(\tau) \left[ \sigma_- \rho \sigma_+ - \frac{1}{2} \{ \sigma_+ \sigma_-, \rho \} \right] \\ & - \frac{1}{4} [k_1(\tau) - 2f_R(\tau)] [\sigma_z \rho \sigma_z - \rho], \end{aligned} \quad (54)$$

where we have set

$$\hat{k}_1(u) = \frac{1 - u\hat{z}(u)}{\hat{z}(u)}. \quad (55)$$

For the case of a single mode the correlation function considered in Eq. (42) explicitly becomes

$$f^1(t) = g^2 e^{i\Delta t}, \quad (56)$$

where the superscript again stresses the fact that a single mode is considered. The solution of the integrodifferential Eq. (41) is then exactly given by the function  $G^1(t)$  introduced in Eq. (32). As it should be, the equations Eq. (33) and Eq. (35) are obtained from Eq. (44) and Eq. (47) under the replacement  $G(t) \rightarrow G^1(t)$ , which corresponds to the special choice of a single mode bath. At the same time it is clear that the explicit coefficients appearing in Eq. (37) can indeed be expressed using real

and imaginary part of Eq. (56), as well as the inverse Laplace transform of

$$\begin{aligned}\hat{k}_1^1(u) &= \frac{1 - u\hat{z}^1(u)}{\hat{z}^1(u)} \\ &= 2g^2 \frac{u}{u^2 + \Delta^2 + 2g^2},\end{aligned}\quad (57)$$

which is given by

$$k_1^1(\tau) = 2g^2 \cos\left(\sqrt{\Delta^2 + 2g^2}\tau\right). \quad (58)$$

### C. Additional term for thermal bath

It is to be stressed, that the possibility to express all relevant functions appearing in time-convolutionless and Nakajima-Zwanzig master equations with reference to the single correlation function  $f(t)$  is a special feature of the two-level system coupled to the vacuum. Indeed while the results of Sect. III for the vacuum are a special case of the model considered in Sect. IV, the more general situation of a thermal bath can be explicitly considered for the case of the reduced dynamics of the Jaynes-Cummings model, showing that the expectation values of various operators as in Eq. (13) have to be specified in order to give the exact equations of motion.

In Sect. III we have seen that the time-convolutionless generator has a different operator structure with respect to the Nakajima-Zwanzig memory kernel only for the case of the vacuum, as it appears comparing Eq. (33) and Eq. (37), while this is no more true for a thermal state. This strongly suggests that the asymmetry in the operator structure of Eq. (44) and Eq. (54) is also due to this special choice of the bath state. To check this fact, in the absence of the exact time evolution map, one has to calculate the time-convolutionless master equation relying on the standard perturbative technique [2], considering terms up to fourth order. The necessity to go up to the fourth perturbative order is immediately clear looking at the interaction Hamiltonian Eq. (39), and observing that the dephasing term, as it appears from Eq. (29), involves a quadrilinear contribution in the raising and lowering operators  $\sigma_+$  and  $\sigma_-$ . This task has been accomplished in Appendix A, leading to the result

$$\begin{aligned}\mathcal{K}_{\text{TCL}}^D(t)\rho &= i\text{Im}\gamma_s(t)[\sigma_+\sigma_-, \rho] \\ &+ \gamma_+(t)\left[\sigma_+\rho\sigma_- - \frac{1}{2}\{\sigma_-\sigma_+, \rho\}\right] \\ &+ \gamma_-(t)\left[\sigma_-\rho\sigma_+ - \frac{1}{2}\{\sigma_+\sigma_-, \rho\}\right] \\ &+ \frac{1}{4}\gamma_d(t)[\sigma_z\rho\sigma_z - \rho],\end{aligned}\quad (59)$$

and the detailed expression of the various coefficients in terms of two- and four-points correlation functions of the system can be found in Eq. (A22) of Appendix A.

This result shows that indeed the disappearance of the dephasing term in the time-convolutionless master equation for the vacuum is a very special feature of this choice of bath. Once again the operator structure of non-Markovian equations of motion is strongly dependent on the details of both bath and interaction term.

## V. CONCLUSIONS

We have obtained the exact time-convolutionless, time-local master equation and Nakajima-Zwanzig integrodifferential master equation for a two-level system coupled to a single bosonic mode, considering a generic bath state. This has been possible thanks to the knowledge of the exact dynamics, and corresponds to the result which can be obtained resumming all terms in the corresponding perturbative techniques. The path followed here, to associate matrices to maps and to consider the direct relation between time evolution map and time local generator, as well as memory kernel, is however much more straightforward. The result shows that in a realistic model each operator contribution in Lindblad form has its own time dependent function, responsible for the non-Markovian behavior, so that a simple multiplication or convolution of the Markovian result with a single phenomenologically guessed function will generally not work. Furthermore the operator structures of the equations of motion in the two cases can strongly differ, also depending on the state of the bath. In particular it has been shown that a dephasing term, which is always present in the Nakajima-Zwanzig equations of motion, disappears for the time-convolutionless case for a bath in the vacuum state. This has been checked for the undamped two-level system thanks to the exact solution, and for the damped case by calculating the fourth order contribution to the time-convolutionless perturbation expansion.

The pursued approach is quite straightforward, even though to obtain the exact expressions it relies on the knowledge of the full time evolution, which can obviously only be feasible for exceptional cases. The detailed analysis of such cases however proves quite useful in understanding the basic features of a non-Markovian description, in particular it puts into evidence the strict relationship between the different quantities which appear in the evolution equations, showing that phenomenological ansatz are in general not easily feasible. Furthermore, the connection to the microscopic perturbative derivation techniques as outlined here, can help in determining the operator structure of the dynamical equations. This structure can sometimes be unveiled calculating the first perturbative contributions, thus restricting the number of ansatz necessary in order to consider a sound phenomenological model.

The necessity to consider non-Markovian equations of motion is often due to strong coupling effects between system and reservoir. This in turn implies that a factorized initial state is not always the most appropriate



initial condition, so that the study of the dynamics of correlated initial states becomes of great significance. This long standing topic has recently seen important advancements [36–43], and we plan to address it in future research work, looking in simple but realistic models, such as the one considered in this article, for general signatures of the effect of initial correlations. These results might help to consider, by means of some approximation, more general systems.

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## Appendix A

We here consider the contributions up to fourth order of the time-convolutionless projection operator technique for the damped two-level system considered in Sect. IV. We assume for the environment an equilibrium state  $\rho_E$ , which commutes with the number operator, and we use the standard projection operator

$$\mathcal{P}w = \text{Tr}_E(w) \otimes \rho_E, \quad (\text{A1})$$

where  $w$  denotes a state of system and environment.

The time-convolutionless perturbative expansion can be obtained by repeated action of the projection operator considered above and of the superoperator

$$\mathcal{L}(t)w = -i[H_I(t), w], \quad (\text{A2})$$

where  $H_I(t)$  is the interaction picture expression corresponding to Eq. (39), given by

$$H_I(t) = \sigma_+(t) \otimes B(t) + \sigma_-(t) \otimes B^\dagger(t), \quad (\text{A3})$$

with

$$\sigma_\pm(t) = e^{\pm i\omega_0 t} \quad (\text{A4})$$

and

$$B(t) = \sum_k g_k b_k e^{-i\omega_k t}. \quad (\text{A5})$$

The contributions to second- and fourth-order for the time-convolutionless generator then read [2]

$$\mathcal{K}_2(t) = \int_0^t dt_1 \mathcal{P}\mathcal{L}(t)\mathcal{L}(t_1)\mathcal{P} \quad (\text{A6})$$

and

$$\begin{aligned} \mathcal{K}_4(t) = & \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 [\mathcal{P}\mathcal{L}(t)\mathcal{L}(t_1)\mathcal{L}(t_2)\mathcal{L}(t_3)\mathcal{P} - \mathcal{P}\mathcal{L}(t)\mathcal{L}(t_1)\mathcal{P}\mathcal{L}(t_2)\mathcal{L}(t_3)\mathcal{P} \\ & - \mathcal{P}\mathcal{L}(t)\mathcal{L}(t_2)\mathcal{P}\mathcal{L}(t_1)\mathcal{L}(t_3)\mathcal{P} - \mathcal{P}\mathcal{L}(t)\mathcal{L}(t_3)\mathcal{P}\mathcal{L}(t_1)\mathcal{L}(t_2)\mathcal{P}] \end{aligned} \quad (\text{A7})$$

respectively. Using these expressions one immediately obtains the operator form of the contributions to the time-convolutionless master equation for the reduced dynamics according to

$$\mathcal{K}_{\text{TCL}}^{(2)}(t)\rho = \text{Tr}_E \left\{ \int_0^t dt_1 \mathcal{L}(t)\mathcal{L}(t_1)\rho \otimes \rho_E \right\} \quad (\text{A8})$$

and

$$\begin{aligned} \mathcal{K}_{\text{TCL}}^{(4)}(t)\rho = & \text{Tr}_E \left\{ \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \right. \\ & [\mathcal{P}\mathcal{L}(t)\mathcal{L}(t_1)\mathcal{L}(t_2)\mathcal{L}(t_3)\mathcal{P}\rho \otimes \rho_E - \mathcal{P}\mathcal{L}(t)\mathcal{L}(t_1)\mathcal{P}\mathcal{L}(t_2)\mathcal{L}(t_3)\mathcal{P}\rho \otimes \rho_E \\ & \left. - \mathcal{P}\mathcal{L}(t)\mathcal{L}(t_2)\mathcal{P}\mathcal{L}(t_1)\mathcal{L}(t_3)\mathcal{P}\rho \otimes \rho_E - \mathcal{P}\mathcal{L}(t)\mathcal{L}(t_3)\mathcal{P}\mathcal{L}(t_1)\mathcal{L}(t_2)\mathcal{P}\rho \otimes \rho_E] \right\}. \end{aligned} \quad (\text{A9})$$

We recall that such high order contributions are needed in order to check the appearance, for an environmental state different from the vacuum, of the dephasing term  $\sigma_z \rho \sigma_z - \rho$ , which involves expressions with altogether four raising and lowering operators of the two-level system.

The second order contribution Eq. (A8) can be expressed by means of the following two correlation functions:

$$\begin{aligned} f(t - t_1) &= e^{i\omega_0(t-t_1)} \text{Tr}_E \{ B(t) B^\dagger(t_1) \rho_E \} \\ &= \sum_k |g_k|^2 e^{i(\omega_0 - \omega_k)(t-t_1)} \langle n_k + 1 \rangle_E, \end{aligned} \quad (\text{A10})$$

which corresponds to Eq. (42) if the bath is in the vacuum state, and

$$\begin{aligned} g(t - t_1) &= e^{-i\omega_0(t-t_1)} \text{Tr}_E \{ B^\dagger(t) B(t_1) \rho_E \} \\ &= \sum_k |g_k|^2 e^{-i(\omega_0 - \omega_k)(t-t_1)} \langle n_k \rangle_E, \end{aligned} \quad (\text{A11})$$

which vanishes in the vacuum. In terms of these functions one has

$$\begin{aligned} \mathcal{P}\mathcal{L}(t_\alpha)\mathcal{L}(t_\beta)\mathcal{P}\rho \otimes \rho_E &= -[f(t_\alpha - t_\beta)\sigma_+\sigma_-\rho + f^*(t_\alpha - t_\beta)\rho\sigma_+\sigma_- \\ &\quad + g(t_\alpha - t_\beta)\sigma_-\sigma_+\rho + g^*(t_\alpha - t_\beta)\rho\sigma_-\sigma_+ \\ &\quad - 2\text{Re} f(t_\alpha - t_\beta)\sigma_-\rho\sigma_+ - 2\text{Re} g(t_\alpha - t_\beta)\sigma_+\rho\sigma_-] \otimes \rho_E. \end{aligned} \quad (\text{A12})$$

This result is sufficient to obtain the time-convolutionless master equation up to second order, indeed upon inserting Eq. (A12) in Eq. (A8) one obtains

$$\begin{aligned} \mathcal{K}_{\text{TCL}}^{(2)}(t)\rho &= -i[\mathfrak{f}_I(t) + \mathfrak{g}_I(t)] [\sigma_+\sigma_-, \rho] \\ &\quad + 2\mathfrak{f}_R(t) \left[ \sigma_+\rho\sigma_- - \frac{1}{2} \{ \sigma_-\sigma_+, \rho \} \right] \\ &\quad + 2\mathfrak{g}_R(t) \left[ \sigma_-\rho\sigma_+ - \frac{1}{2} \{ \sigma_+\sigma_-, \rho \} \right], \end{aligned} \quad (\text{A13})$$

where we have set

$$\mathfrak{f}(t) = \int_0^t dt_1 f(t - t_1) \quad (\text{A14})$$

and

$$\mathfrak{g}(t) = \int_0^t dt_1 g(t - t_1), \quad (\text{A15})$$

denoting as usual real and imaginary parts with the subscripts  $R$  and  $I$  respectively.

To consider the fourth order contribution one has to evaluate the four terms given in Eq. (A9). The last three terms at the right hand side can be obtained applying twice the result Eq. (A12), thus obtaining

$$\begin{aligned} \mathcal{P}\mathcal{L}(t)\mathcal{L}(t_\alpha)\mathcal{P}\mathcal{L}(t_\beta)\mathcal{L}(t_\gamma)\mathcal{P}\rho \otimes \rho_E &= [-4\sigma_+\rho\sigma_- \{ \text{Re} f(t - t_\alpha) \text{Re} g(t_\beta - t_\gamma) + \text{Re} g(t - t_\alpha) \text{Re} g(t_\beta - t_\gamma) \} \\ &\quad - 4\sigma_-\rho\sigma_+ \{ \text{Re} g(t - t_\alpha) \text{Re} f(t_\beta - t_\gamma) + \text{Re} f(t - t_\alpha) \text{Re} f(t_\beta - t_\gamma) \} \\ &\quad + \sigma_+\sigma_-\rho f(t - t_\alpha) f(t_\beta - t_\gamma) + \rho\sigma_+\sigma_- f^*(t - t_\alpha) f^*(t_\beta - t_\gamma) \\ &\quad + \sigma_-\sigma_+\rho g(t - t_\alpha) g(t_\beta - t_\gamma) + \rho\sigma_-\sigma_+ g^*(t - t_\alpha) g^*(t_\beta - t_\gamma) \\ &\quad + \sigma_+\sigma_-\rho\sigma_+\sigma_- \{ 2\text{Re} [f(t - t_\alpha) f^*(t_\beta - t_\gamma)] \\ &\quad + 4\text{Re} g(t - t_\alpha) \text{Re} f(t_\beta - t_\gamma) \} \\ &\quad + \sigma_-\sigma_+\rho\sigma_-\sigma_+ \{ 2\text{Re} [g(t - t_\alpha) g^*(t_\beta - t_\gamma)] \\ &\quad + 4\text{Re} f(t - t_\alpha) \text{Re} g(t_\beta - t_\gamma) \} \\ &\quad + \sigma_-\sigma_+\rho\sigma_+\sigma_- \{ f^*(t - t_\alpha) g(t_\beta - t_\gamma) + g(t - t_\alpha) f^*(t_\beta - t_\gamma) \} \\ &\quad + \sigma_+\sigma_-\rho\sigma_-\sigma_+ \{ g^*(t - t_\alpha) f(t_\beta - t_\gamma) + f(t - t_\alpha) g^*(t_\beta - t_\gamma) \}] \otimes \rho_E, \end{aligned} \quad (\text{A16})$$

where the relations  $\sigma_+^2 = \sigma_-^2 = 0$  have been repeatedly used, together with the assumption  $[\rho_E, n_k] = 0$ .

The first term at the right hand side of Eq. (A9) instead requires the introduction of a four-point correlation function, which is given by

$$h(t_a, t_b, t_c, t_d) = e^{i\omega_0(t_a - t_b + t_c - t_d)} \text{Tr}_E \{ B(t_a) B^\dagger(t_b) B(t_c) B^\dagger(t_d) \rho_E \}, \quad (\text{A17})$$

with  $B(t)$  as in Eq. (A5). An explicit evaluation of  $\mathcal{P}\mathcal{L}(t)\mathcal{L}(t_1)\mathcal{L}(t_2)\mathcal{L}(t_3)\mathcal{P}\rho \otimes \rho_E$  together with the repeated use of Eq. (A16) then leads to the desired result, which can be obtained with a straightforward though very lengthy calculation. The fourth order contribution reads

$$\begin{aligned} \mathcal{K}_{\text{TCL}}^{(4)}(t)\rho = & i [\mathbf{p}_I(t) + \mathbf{r}_I(t) + \mathbf{v}_I(t)] [\sigma_+ \sigma_-, \rho] \\ & + \mathbf{t}(t) \left[ \sigma_+ \rho \sigma_- - \frac{1}{2} \{ \sigma_- \sigma_+, \rho \} \right] \\ & + \mathbf{u}(t) \left[ \sigma_- \rho \sigma_+ - \frac{1}{2} \{ \sigma_+ \sigma_-, \rho \} \right] \\ & + \frac{1}{4} [\mathbf{q}(t) + \mathbf{s}(t) + 2\mathbf{v}_R(t)] [\sigma_z \rho \sigma_z - \rho], \end{aligned} \quad (\text{A18})$$

where in analogy to the notation of Eq. (A14) and Eq. (A15) we use the Fraktur character to denote the triple integral over time of the function with the corresponding Roman letter, for example

$$\mathbf{p}(t) = \int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 p(t, t_1, t_2, t_3). \quad (\text{A19})$$

The functions determining the coefficients appearing in Eq. (A18) are given in terms of the above introduced two- and four-points correlation functions of the model according to the expressions

$$\begin{aligned} p(t, t_1, t_2, t_3) &= - \sum_{\alpha\beta\gamma} f(t - t_\alpha) f(t_\beta - t_\gamma) + h(t, t_1, t_2, t_3) \\ q(t, t_1, t_2, t_3) &= -2 \sum_{\alpha\beta\gamma} \{ \text{Re}[f(t - t_\alpha) f^*(t_\beta - t_\gamma)] + 2 \text{Re} g(t - t_\alpha) \text{Re} f(t_\beta - t_\gamma) - \text{Re} h(t_\alpha, t, t_\beta, t_\gamma) \} \\ r(t, t_1, t_2, t_3) &= g(t - t_2) g(t_1 - t_3) + g(t - t_3) g(t_1 - t_2) + f(t_1 - t) f(t_3 - t_2) - h(t_1, t, t_3, t_2) \\ s(t, t_1, t_2, t_3) &= -2 \sum_{\alpha\beta\gamma} \{ \text{Re}[f(t - t_\alpha) f(t_\gamma - t_\beta)] + 2 \text{Re} f(t - t_\alpha) \text{Re} g(t_\beta - t_\gamma) - \text{Re} h(t, t_\alpha, t_\gamma, t_\beta) \} \\ t(t, t_1, t_2, t_3) &= 2 \sum_{\alpha\beta\gamma} \{ \text{Re}[f(t - t_\alpha) f(t_\gamma - t_\beta)] + \text{Re}[g(t - t_\alpha) g(t_\beta - t_\gamma)] \\ &\quad + 2 \text{Re} f(t - t_\alpha) \text{Re} g(t_\beta - t_\gamma) - \text{Re} h(t, t_\alpha, t_\gamma, t_\beta) \} \\ &\quad + 2 \{ \text{Re}[f(t_1 - t) f(t_3 - t_2)] - \text{Re}[g(t - t_1) g(t_2 - t_3)] - \text{Re} h(t_1, t, t_3, t_2) \} \\ u(t, t_1, t_2, t_3) &= 2 \sum_{\alpha\beta\gamma} \{ \text{Re} f(t - t_\alpha) \text{Re} f(t_\beta - t_\gamma) + 2 \text{Re} g(t - t_\alpha) \text{Re} f(t_\beta - t_\gamma) - \text{Re} h(t_\alpha, t, t_\beta, t_\gamma) \} \\ &\quad - 2 \text{Re} h(t, t_1, t_2, t_3) \\ v(t, t_1, t_2, t_3) &= 2 \sum_{\alpha\beta\gamma} \{ f(t_\alpha - t) f(t_\gamma - t_\beta) - h(t_\alpha, t, t_\gamma, t_\beta) \}, \end{aligned} \quad (\text{A20})$$

where the following summation convention has been used

$$\sum_{\alpha\beta\gamma} \psi(t_\alpha, t_\beta, t_\gamma) = \psi(t_1, t_2, t_3) + \psi(t_2, t_1, t_3) + \psi(t_3, t_1, t_2). \quad (\text{A21})$$

Including terms up to fourth order one therefore has the expression Eq. (59) with time dependent coefficients given by the identifications

$$\begin{aligned} \gamma_s(t) &= -\mathbf{f}(t) - \mathbf{g}(t) + \mathbf{p}(t) + \mathbf{r}(t) + \mathbf{v}(t) \\ \gamma_+(t) &= 2\mathbf{f}_R(t) + \mathbf{t}(t) \\ \gamma_-(t) &= 2\mathbf{g}_R(t) + \mathbf{u}(t) \\ \gamma_d(t) &= \mathbf{q}(t) + \mathbf{s}(t) + 2\mathbf{v}_R(t). \end{aligned} \quad (\text{A22})$$

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